Bis[4-(4-maleimidephen-oxy)phenyl]propane/N,N-4,4′-Bismaleimido-diphenylmethyene Blend Modified with Diallyl Bisphenol A

Chunyan Qu,[1] Liwei Zhao,[1] Dezhi Wang,[1] Hongfeng Li,[1] Wanbao Xiao,[1] Cheng'e Yue[2]

[1]Institute of Petrochemistry, Hei Long Jiang Academy of Sciences, Harbin 150040, People's Republic of China
[2]College of Materials Science and Engineering, Qiqihar University, Qiqihar 161006, People's Republic of China
Correspondence to: C. Qu (E-mail: quchunyan168@163.com)

ABSTRACT: In this article, 2,2′-bis[4-(4-maleimidephen-oxy)phenyl)]propane (BMPP) resin and N,N-4,4′-bismaleimidodiphenyl-methyene (BDM) resin blends were modified by diallyl bisphenol A (DABPA). The effects of the mole concentration of BMPP on mechanical properties, fracture toughness, and heat resistance of the modified resins were investigated. Scanning electron microscopy was used to study the microstructure of the fractured modified resins. The introduction of BMPP resin improves the fracture toughness and impact strength of the cured resins, whose thermal stabilities are hardly affected. Dynamic mechanical analysis shows that the modified resins can maintain good mechanical properties at 270.0°C, and their glass transition temperatures (T_g) are above 280.0°C. When the mole ratio of BDM : BMPP is 2 : 1(Code 3), the cured resin performs excellent thermal stability and mechanical property. Its T_g is 298°C, and the Charpy impact strength is 20.46 KJ/m^2. The plane strain critical stress intensity factor (K_{IC}) is 1.21 MPa·m$^{0.5}$ and the plane strain critical strain energy release rate (G_{IC}) is 295.64 J/m^2. Compared with that of BDM/DABPA system, the K_{IC} and G_{IC} values of Code 3 are improved by 34.07% and 68.10%, respectively, which show that the modified resin presented good fracture toughness. © 2014 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 40395.

INTRODUCTION

Bismaleimide (BMI) resins have excellent properties, such as high temperature resistance, chemical resistance, moisture resistance, high glass transition temperature, and excellent mechanical properties.[1–5] Therefore, BMI resins can be widely used as advanced composite matrix, wear-resistant material, high temperature-resistant insulation material, high-temperature structural adhesive in aerospace, electronic products, and transportation.[3,6] However, BMI resins, such as N,N-4,4′-bismaleimidodiphenylmethyene (BDM), are hard to process[7] for their high mold temperature, and the cured products are brittle because of the high cross-linking densities and short-chain length in the backbone,[8] which limits their structural applications and developments. So, lots of works have been performed to achieve good fracture toughness of the BMI resins. One way is to use diamines, dithiols, and allylphenols such as 2,2′-diallyl bisphenol A (DABPA) as chain extenders in BMI resins to decrease the cross-linking densities of the cured resins.[1,9,10] Engineering thermoplastics[7,11,12] such as polyamide-imide, polyether ketone ketone, and reactive liquid rubber[13–15] (e.g., carboxyl-terminated nitrile rubber[10] and vinyl nitrile-terminated rubber) are used as tougheners for modifying the BMI resins. However, they usually decrease the thermal stability of the

cured resins. Inorganic nanoparticles[16–18] and whiskers are also used as flexibilizers to enhance fracture toughness of BMI resins. In order to enhance the toughness of BMI resins without sacrificing the excellent thermal properties, some long-chain structure BMI resins are synthesized.[1,4,19] These BMI resins showed well performance on the mechanical properties, especially the fracture toughness of the cured resins, without significant reducing the heat resistance of the modified resins.[15]

In this article, a mixed BMI resin system consisting of BDM, bis[4-(4-maleimidephen-oxy)phenyl)]propane (BMPP), and DABPA were studied. The mechanical properties, fracture toughness, and heat resistance of the modified resins were investigated.
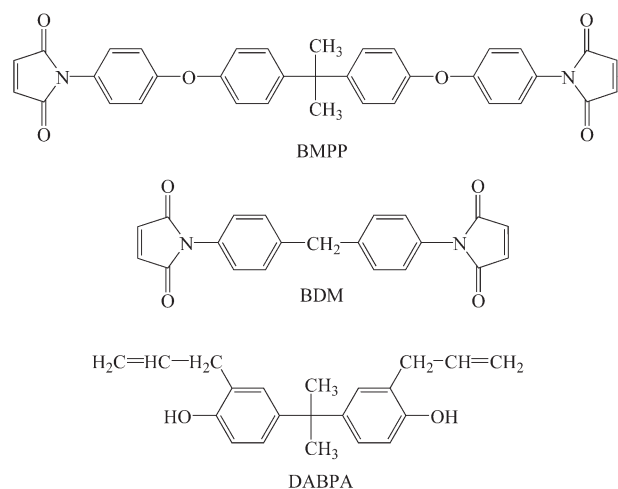
EXPERIMENTAL

Materials
DABPA, BDM, and BMPP are commercial products. Their chemical structures are shown in Figure 1.

Preparations of BMPP/BDM/DABPA Blends and the Cured Resins
The DABPA and BDM/BMPP were weighed, respectively, following Table I, and then they were put into a reaction flask.

Figure 1. Chemical structure of BMPP, BDM, and DABPA.

Following, they were mixed with continuous stirring for 30 min at 130–135°C to form the prepolymer, and degassed in a vacuum oven at 120–125°C for 1.5 h. And then the low viscosity resin was poured into the mold release agent-coated metal molds. Followed the curing reaction was carried out via the procedure of 165°C/2 h + 180°C/2 h + 200°C/2 h + 238°C/4 h, and postcuring for 5 h at 250°C in an air-circulated oven. The oven was then turned off and cooled to room temperature slowly, and then the cured resin bars were taken out from molds.

Differential Scanning Calorimetry Analysis
Differential scanning calorimetry (DSC) tests of the prepolymer were performed on DSC6220 in a nitrogen atmosphere at a 50 mL/min flow rate. A heating rate of 10°C/min was applied from 25°C to 400°C.

Rheology Studies
In this article, A Gemini 200 rheometer was used to study the rheological property of the prepolymer. Samples were prepared by pressing about 1.5 g resins into 1.3 mm thick disks. A frequency of 1 Hz and a temperature ramping rate of 4°C/min were used during test. Then the melt viscosities and gel point of the prepolymer during the cross-linking process were recorded.

Mechanical Properties
Tensile test and three-point bend (3PB) test of the cured resins were performed on Instron 4467 universal testing machine per ASTM D 638-10 and ASTM D 790-10, respectively.[20,21] Size of

3PB test specimen was (80 ± 0.2) mm $\times$ (15 ± 0.2) mm $\times$ (4.0 ± 0.2) mm. Charpy impact test was accomplished per ASTM D 6110-06 on ZBC7000, and the specimens size were 127 mm $\times$ 12.7 mm $\times$ 6.35 mm.[22] At least five specimens were used for the test above.

Fracture Toughness
The single-edge notch 3PB (SEN-3PB) test was accomplished per ASTM D 5045-99 using universal testing machine (Instron 4467) to calculate the plane strain critical stress intensity factor (K_{IC}), and the specimens size were 127 $\times$ 12.7 $\times$ 6.35 mm^3.[23–25] The SEN-3PB bars were notched with a milling tool (250 μm tip radius), followed by liquid nitrogen-chilled razor blade tapping to wedge open a shark crack with a parabolic crack front.[13,25] The ration of the crack length (a) and the specimen width (W) was chosen to be between 0.45 and 0.55 for the SEN-3PB test. A constant crosshead speed of 10 mm/min was chosen to conduct SEN-3PB test. At least ten SEN-3PB specimens were used for the K_{IC} measurement.

The K_{IC} value was calculated using the following equation:

$$K_{IC} = \frac{P_Q \cdot f(x)}{B \cdot W^{1/2}} \tag{1}$$

Here $f(x)$ is given by the following equation from ASTM D 5045-99:

$$f(x) = 6x^{\frac{1}{2}} \frac{[1.99 - x(1-x)(2.15 - 3.93x + 2.71x^2)]}{(1+2x)(1-x)^{\frac{3}{2}}} \tag{2}$$

Here B and W are the thickness and width of the SEN-3PB specimen. The x value is equal to a/W. The P_Q value in eq. (1) is equal to the maximum load because that all specimens exhibited the brittle fracture behavior.

The plane strain critical strain energy release rate (G_{IC}) can be calculated by the following relationship:

$$G_{IC} = \frac{K_{IC}^2 \cdot (1 - v^2)}{E} \tag{3}$$

Here E is the Young's modulus and v is the Poisson's ratio. In this article, v is assumed to be 0.36 for all modified resins.

Dynamic Mechanical Analysis
The dynamic mechanical behavior of the modified BMI resins was studied by DMS6100 in a three-point bending mode with a 5°C/min heating rate. A constant strain amplitude of 1% and a frequency of 1 Hz were used. The samples were tested at temperatures ranging from 25°C to 350°C, and tan δ peak was assigned as T_g.

Table I. Formulas of the Blends

Code	BDM : BMPP (mole ratio)	Average molecular weight of BDM/BMPP	BDM/BMPP : DABPA (mole ratio)	BDM/BMPP : DABPA (weight ratio)
0	1 : 0	358	1 : 0.87	100 : 75
1	1 : 1	464.30	1 : 0.87	100 : 56.96
2	1.5 : 1	443.04	1 : 0.87	100 : 59.70
3	2 : 1	428.87	1 : 0.87	100 : 61.67
4	2.5 : 1	418.74	1 : 0.87	100 : 63.16
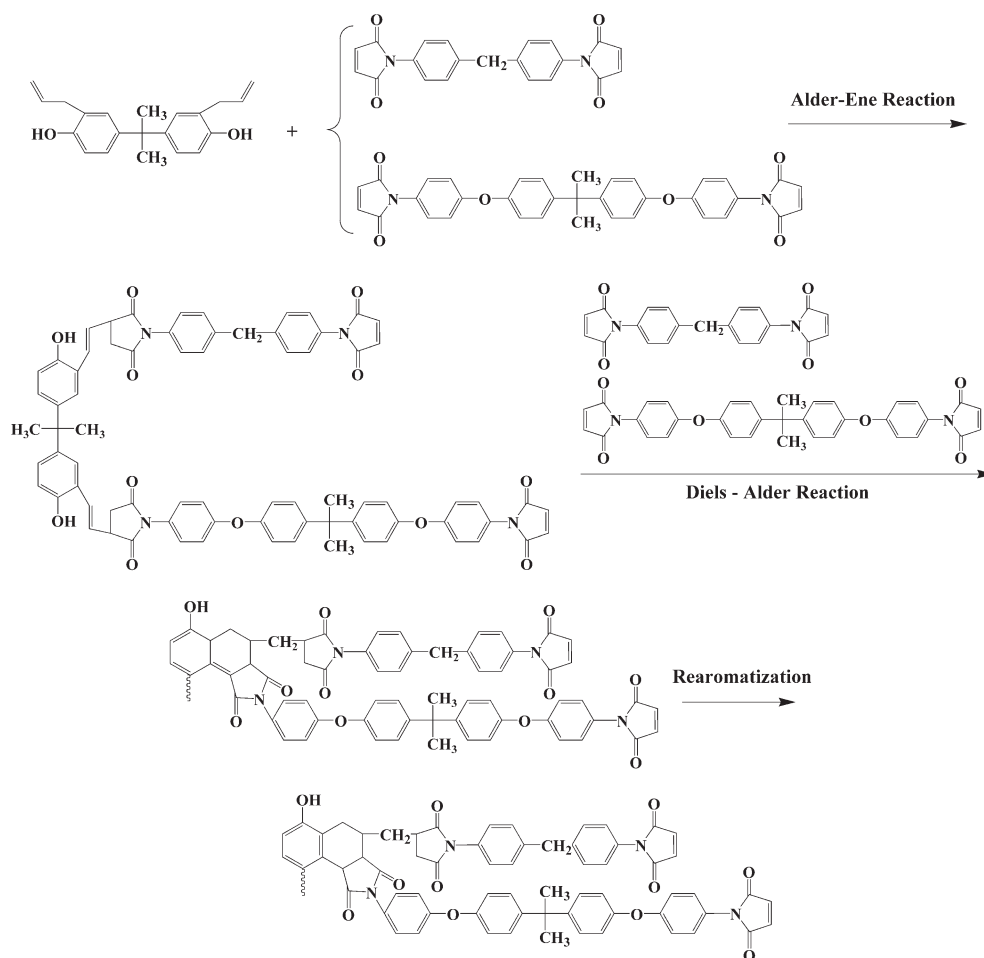
Figure 2. The main reaction scheme of the BDM/BMPP/DABPA system.

Thermogravimetric Analysis

Thermogravimetric analyses (TGAs) of the cured resins were carried out using DTA6300 with a heating rate of 5°C/min in a nitrogen atmosphere. And the 5% loss weight temperature (T_{d5}) was assigned as the initial degradation temperature (T_{di}).[3] The temperature at maximum degradation rate was collected with DTA method.

Morphology of the Fracture Surface

Scanning electron microscopy (SEM, FEI Sirion) was used to study the fracture surface of the specimens. The detailed microscopy procedures for preparing the samples can be found in the literatures.

RESULTS AND DISCUSSION

Reaction Mechanism

The curing reaction between allyl bisphenol A and BMI monomer has been established by the previous studies. First, the C=C groups in BMI molecule react with the C=C groups in DABPA molecule, and then the intermediates are generated by Alder–Ene reaction. Second, the Diels–Alder reaction carries out under a high temperature,[26,27] while the reaction process of BDM/BMPP/DABPA system is shown in Figure 2.

Rheology Characterization

Figure 3 shows that the melt viscosities of the blends change with the temperature increases. It can be seen that, compared with BDM/DABPA system, BDM/BMPP/DABPA system can maintain low melt viscosity in a wide temperature range. The
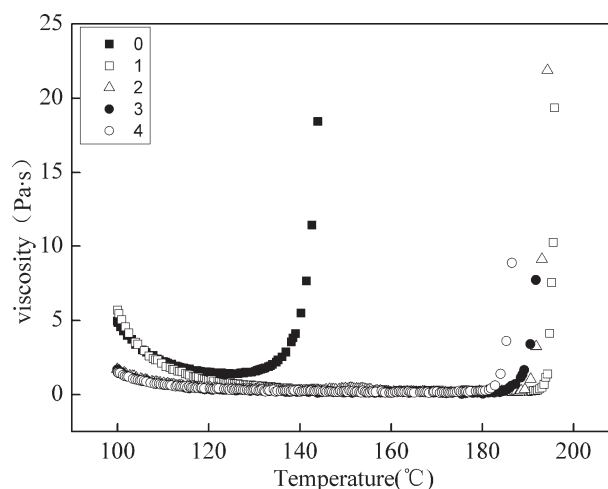
Figure 3. Viscosity–temperature curves of the prepolymer.
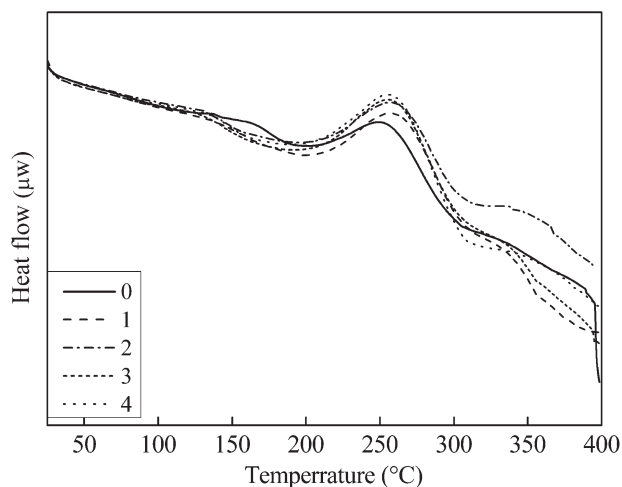
Figure 4. DSC curves of the prepolymer.

ternary blending system can keep a low viscosity of <1 Pa·s when the temperature ranges from 120°C to 180°C. The viscosity of BDM/DABPA system decreases with temperature increasing until the temperature reached 125°C, after which, the viscosity increases rapidly as the temperature increases. However, the melt viscosity of the BDM/BMPP/DABPA system does not increase quickly until the temperature exceeds 180°C. That demonstrates that BMPP can effectively broaden the processing window of the modified resin. The introduction of BMPP in the modified resin system increases the flow property of the blends in a wide temperature range, which is good to process for the modified resin. The low melt viscosity of the mixed resin is suitable for the resin transfer molding (RTM), which need the wide processing range of temperature and low injection pressure.[10]

DSC Characterization

The exothermic situation of the modified resin during curing reaction was characterized by DSC (Figure 4). BMPP affects slightly the exothermic peak of the modified resin system. The curing temperature at peak top (T_P) of the BDM/DABPA system is 252.6°C while that of the Code 3 is 258.9°C. The concentration of BMPP is greater, the T_P is higher, and that is the same with the initial and the finishing curing temperature. The molecular chain between the two maleimide groups is long in the chemical structure of BMPP compared with that in BMI, which decreases the molar ratio of maleic anhydride in the main chain and elongates the distance of the reaction point reacting with the allyl groups in DABPA.[1,26] Therefore, the apparent activation energy (E_a) during curing reaction become higher and the curing temperatures is increased.

In this article, prepolymer are used in rheological and DSC tests as samples. The blends were mixed with continuous stirring for 30 min at 130–135°C to form the prepolymer. Under the above condition, the C=C groups in BMI molecule can react with the C=C groups in DABPA molecule through Alder–Ene reaction and the exotherms do not show up in DSC curves. The molecular chain between reacting sites is shorter in BDM molecule than that in BMPP molecule. That BDM reacts with DABPA is

much easier, and the viscosity of BDM/DABPA system increases rapidly at about 140°C when the prepolymer is put into a rheometer and heated again. Compared with system without BMPP, the total apparent activation energy (E_a) of system with BMPP becomes higher. The ternary system can keep low viscosity at high temperature as a consequence.

A three-dimensional network structure of the system is formed through Diels–Alder reaction at about 180°C for all systems in this article. Viscosity of the modified system shows a dramatic increase because of the existence of cross-linking structure. This can be seen in Figure 3.

Exotherms begin to show a big increase at about 180°C as the three-dimensional network structure is formed. Therefore, there is relatively little difference in the exotherms for the resins in DSC curves.

Mechanical Properties

Stress-strain curves of the cured modified resins are given in Figure 5. The curves shows that strain increases as stress increases for all cured resins. Fracture strains of the cured resins are 0.03200 mm/mm, 0.03155 mm/mm, 0.03202 mm/mm, 0.03318 mm/mm, and 0.03381 mm/mm for Codes 0, 1, 2, 3, and 4, respectively. Strain of different cured resin is nearly the same at the same stress when stress value is below 24 MPa. The strain increase differently when the stress exceeds 24 MPa. The cured resin shows a big strain increase with the same stress increase because of low cross-linking destiny and introduction of ether group.

Tensile strength and bend strength of the cured resin are two important indicators when characterizing the mechanical properties of materials. The tensile strength and tensile modulus of the modified resins are shown in Figure 6, and the bend strength and bend modulus of different systems are shown in Figure 7. BMPP decreases the tensile strength and tensile modulus slightly. Compared with the other formulas, the tensile strength drops obviously when the concentration of BMPP increases. However, tensile modulus of all modified systems is hardly affected. So the rigidity of modified cured resin is kept. It can be seen that the introduction of BMPP increases the bend strength of
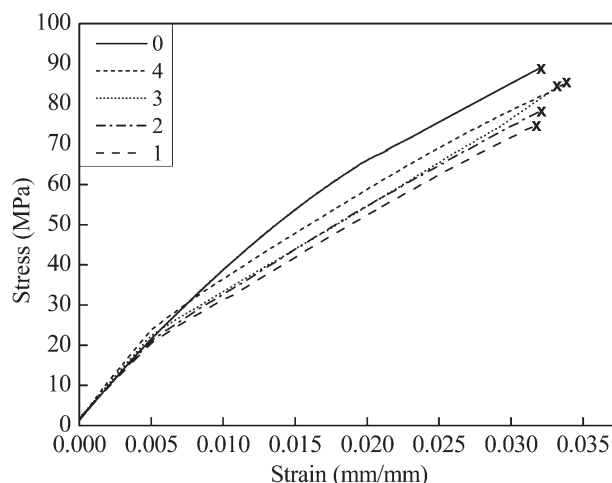

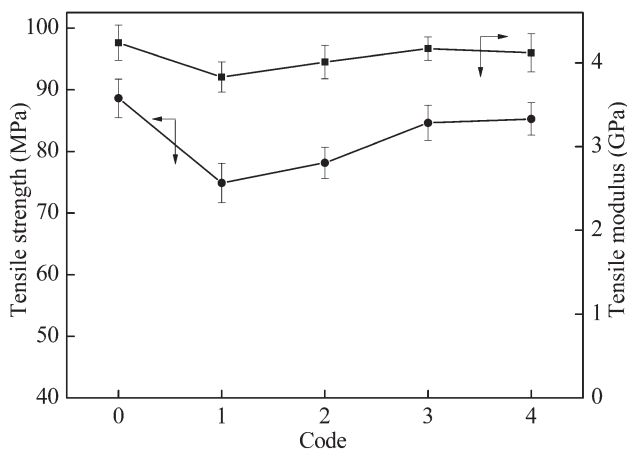**Figure 5.** Stress–strain curves of the cured resins.

Figure 6. Tensile strengths and tensile moduli of the cured resins.

Figure 8. Impact strengths of the cured resins.

the modified resins. The bend strength of BDM/DABPA is 152.33 MPa. When the mole ratio of BDM : BMPP is 2 : 1, the bend strength of the system reaches its maximum of 170.40 MPa. It means that the bend strength increases by 11.87% with maintaining the same bend modulus. The bend modulus and tensile modulus of classical XU292 BMI resin system are 3.98 GPa and 3.90 GPa, respectively. While that of the Code 3 system in this article are 4.11 GPa and 4.17 GPa, respectively, which means that the cured resin (Code 3) has good properties. So the introduction of moderate BMPP into the modified system increases the bend strength of the cured resin without decrease its tensile strength almost. Moreover, it is seen in dynamic mechanical analysis traces (Figure 10) that the modified system can maintain a very higher storage modulus at 270.0°C.

Toughness and Fracture Toughness
In this study, Charpy impact strength, the plane strain critical stress intensity factor (K_{IC}) and the plane strain critical strain energy release rate (G_{IC}) were used to characterize the toughness of the modified resins (Figures 8 and 9). The impact test result shows that BMPP increase the toughness by a large degree. The impact strength of BDM/DABPA system is 10.13 KJ/m^2, which indicates that the cured resin is brittle. The impact strength of the modified resin reaches 20.46 KJ/m^2 when the mole ratio of BDM : BMPP is 2 : 1 in the modified
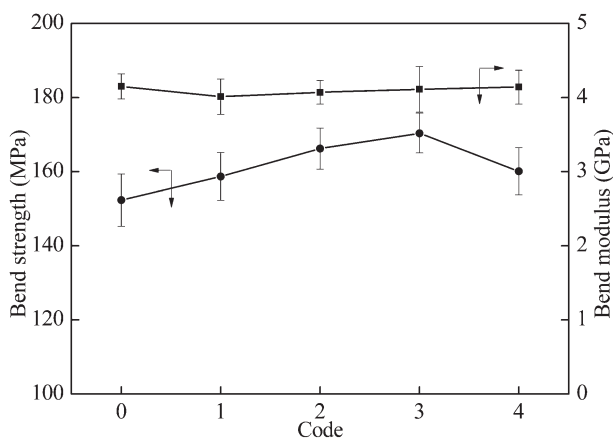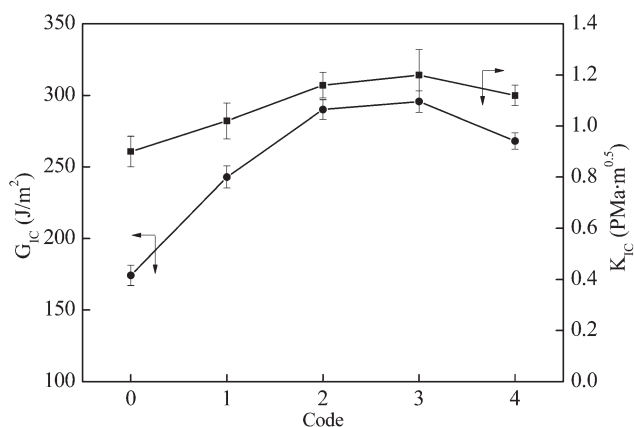

Figure 9. K_{IC} and G_{IC} values of the cured resins.

system. It can be seen in Figure 9 that the K_{IC} and G_{IC} values increase greatly. The K_{IC} and G_{IC} values of the cured resin reach their maximum values when the concentration of BMPP is the same. The K_{IC} maximum value is 1.21 MPa·$m^{0.5}$ and the G_{IC} maximum value is 295.64 J/m^2. The values are enhanced by 34.07% and 68.10%, respectively, compared with that of BDM/DABPA system. It is obvious that moderate BMPP largely improves the toughness of the modified system.

The average molecular weight between cross-links (Mc) in the cured BMPP molecule is bigger than that of the common cured BDM resin, which can decrease the cross-linking density of the cured resin. The toughness of the cured resin increases when the cross-linking density decreases in a degree. The flexible ether bonds introduced by BMPP can reduce brittleness of the cured

Figure 7. Bend strengths and bend moduli of the cured resins.

Table II. Fracture Toughness of Some BMI Resin Systems

	5250-4	AT-PAEKI modified BPA-BMI	Code 3
K_{IC} (MPa·$m^{0.5}$)	0.87	0.55	1.21
G_{IC} (J/m^2)	160	–	295.64
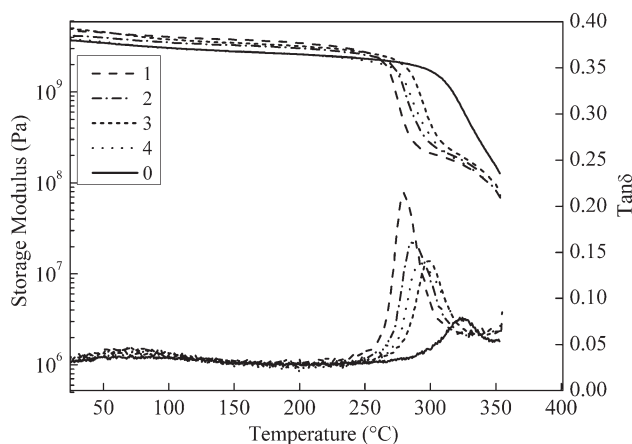
Figure 10. Dynamic mechanical spectra of the modified cured resins.

Figure 11. Thermogravimetric curves of the modified cured resins.

Table III. The Effects on T_gs of the Cured Resins of Ether Group Concentration and M_{AR}

Code	0	1	2	3	4
Ether group concentration (mol/mol)	0	1.0	0.8	0.67	0.57
M_{AR} (g/mol)	166	272	251	237	227
T_g (°C)	324	281	286	298	292

Table IV. T_g Values of Some BMI Resin Systems

	BMI resin systems			
Cytec 5250-4	Cytec 5270	Hexcel F-178	Ciba-Geigy XU292	Code 3
T_g (°C)	295	287	275	287	298 (dry) 288 (wet)[a]

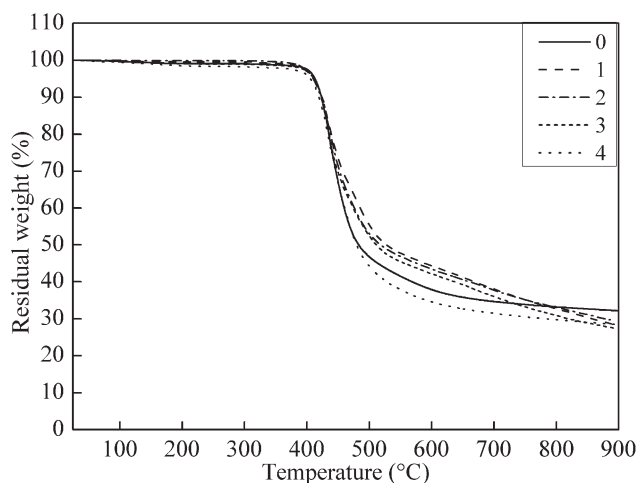[a] The sample was treated under the following conditions: 71°C, 95% humidity for 2 weeks (wet T_g data of Code 3).

Dynamic Mechanical Analysis

The dynamic mechanical spectra of the modified resins are shown in Figure 10. The T_gs of thermosetting resins are very important properties because they are the maximum application temperatures of the resins. The glass transition temperature (T_g) of the BDM/DABPA resin system is 324°C. There is a linear relationship of the T_g values between the BMPP resin and the BDM resin with different quality ratios, excepted the Code 3. T_g of Code 3 is higher than that of Code 4. T_gs of all modified systems are above 280°C and the cured resin has good mechanical property at 270°C. T_g is reduced because of the introduction of BMPP, but the storage modulus of the cured resins is not affected at all until the temperature reaches 250°C.

The effects on T_gs of the ether group concentration and the average molecular weight (M_{AR}) between the two maleimide groups in the cured BMPP resin and the cured BDM are shown in Table III. The ether group mole content per mole BDM/BMPP resin is assigned as ether group concentration. It can be seen from Table III that T_g of the cured resin decreases with the increasing ether group concentration and M_{AR} generally. The chain between cure sites is long in the BMPP molecular, which can decrease the cross-linking density of the cured resin. The ether groups in BMPP molecular also can lower T_g value of the cured resin, because the ether bond is flexible. The flexible ether bond in the cured resin can move easily at not very high temperature, which may cause decreasing in storage modulus and

resin and improve toughness of the cured resin. As a result, the cured resin can absorb more energy before fracture.

The K_{IC} and G_{IC} values of the Code 3 system are remarkable compared with that of Cytec 5250-4-RTM BMI resin (data from Ref. 13) and AT-PAEKI modified BPA-BMI resin system (data from Ref. 8). The K_{IC} and G_{IC} values are shown in Table II.

Table V. T_g and Characteristic Data From TG Curves

Code	T_g (°C)	T_5 (°C)	T_{30} (°C)	Temperature of maximum degradation rate (°C)	Char yield at 600°C (%)	Char yield at 900°C (%)
0	324	412.95	446.35	433.69	37.92	32.17
1	281	411.07	457.60	428.36	44.35	28.22
2	286	410.24	452.45	431.68	43.45	29.25
3	298	411.89	449.37	430.15	42.18	27.26
4	292	405.46	446.04	432.52	34.59	28.43

Figure 12. SEM images of fracture surface of (A) the cured BDM/DABPA resin and (B) the cured BDM/BMPP/DABPA resin (Code 3).

T_g in a degree. However, the reduction is not significant and can be acceptable in many applications.

The cured Code 3 resin has good mechanical properties and excellent toughness and fracture toughness. Moreover, it shows a high glass transition temperature of 298°C, and the storage modulus can maintain 2.33 GPa up to 270°C, which means that the cured resin can keep good mechanical properties at high temperature. That is excellent compared with that of some commercial BMI resin systems. Some T_g values of BMI resins are shown in Table IV.

TGA Analysis of the Cured Resin

The heat resistance of the cured resin was characterized with TGA method and the thermogravimetric curves of the different cured resin system are given in Figure 11. BMPP hardly decrease the initial degradation temperature of the cured resin. The T_{di} of the cured resins is all exceed 400°C. And the temperature of maximum degradation rate is almost not affected, which means that the cured modified resins can hold stability in a high temperature. For Codes 1, 2, and 3, the temperature of losing 30% weight and the char yield at 600°C are even improved. So

BMPP does not cause a significant reduction in thermal stability. Same characteristic data are listed in Table V.

SEM Characterization

The micromorphology of the fracture cured resin surface was characterized through SEM equipment. The test results are shown in Figure 12. The micrograph (Figure 12 A) is the SEM images of the fracture surface of BDM/DABPA. The fracture surface of BDM/DABPA is neat and smooth and in one direction, which indicates a typical brittle fracture mode. The micrograph (Figure 12 B) is for BDM/BMPP/DABPA system (Code 3). The fracture surface is rough and uneven, and the damaging direction is discontinuity. It is a form of ductile fracture. It can be seen from the SEM test that BMPP can enhance the toughness of the modified resin in some degree. The SEM test result is consistent with that of the mechanical test.

CONCLUSIONS

A series of BDM and BMPP blends modified by DABPA were prepared in the research. The rheological property of the pre-polymer, mechanical properties, fracture toughness, and thermal stability of the cured resin were investigated. The results show that the introduction of moderate BMPP into the system can broaden the processing window of the modified resin without affecting the curing temperature of the modified resin, and the blend has a good flow property in a wide temperature range. The BDM/BMPP/DABPA ternary blending system can keep a low viscosity of <1 Pa·s with a temperature range from 120°C to 180°C, which means that the modified resin can wet fiber well when used as resin matrix in fiber composites.

BMPP can improve the toughness, fracture toughness and the bend strength of the cured resin without decreasing mechanical properties and heat resistance significantly. The K_{IC} maximum value is 1.21 MPa·m$^{0.5}$ and the G_{IC} maximum value is 295.64 J/m^2 and the values are enhanced by 34.07% and 68.10%, respectively, compared with that of BDM/DABPA system. The values are remarkable compared with that of some modified BMI system (commercial and reference). T_gs of all modified systems are above 280°C and the cured resin has good mechanical property at 270°C. The cured resin has a high T_g of 298°C when the mole ratio of BDM : BMPP is 2 : 1 in the modified system, and possess good mechanical properties and excellent toughness and fracture toughness. BMPP hardly decrease the initial degradation temperature of the cured resin. The T_{di} of the cured resins are all exceed 400°C in a nitrogen condition and the temperature of maximum degradation rate is almost not affected.

ACKNOWLEDGMENTS

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